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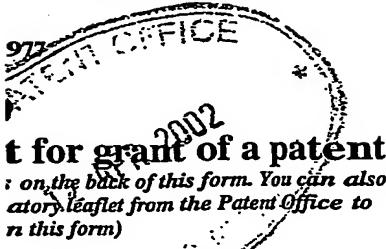
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FILM COATING

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## FILM COATING

The present invention relates to a coating composition for improving the surface smoothness of a substrate on which it is coated, particularly a polymeric substrate for use in electroluminescent (EL) display devices, in particular organic light emitting display (OLED) devices such as polymer light emitting display (PLED) devices.

Electroluminescent (EL) display is a self-emitting display mode which features excellent visibility (including high brightness, high contrast, very fast response speed and wide viewing angle), an extremely thin profile and very low power consumption. The EL display device itself emits light, as do cathode ray tubes (CRT), fluorescent and plasma displays. Consequently, the need for backlighting, which is required for most liquid crystal displays (LCDs), is eliminated. EL displays may be used in a variety of applications, including aircraft and ship controls, automobile audio equipment, calculators, mobile telephones, portable computers, instrumentation, factory monitors and electronic medical equipment. Another major application for EL displays is as a light source, particularly as backlighting for small LCD panels in order to render them easier to read in low ambient light conditions.

EL displays work by sandwiching a thin film of a phosphorescent or other electroluminescent substance between two plates each of which comprises conductive elements in a predetermined pattern, i.e. electrodes, thereby forming addressable pixels on the display. The electrodes are formed as coatings either on the electroluminescent substance or on a separate support. Where the or each electrode is intended to transmit light, the electrodes are formed as translucent or transparent coatings, for instance using transparent conductive metal oxides. Equally, the or each support may be translucent or transparent, as required. Generally, at least the anode is transparent. The support generally functions both as a base for an electrode and as an insulating layer. The substrate also provides protection against chemical and physical damage in use, storage and transportation. Glass, as well as polymeric film, has been used as the insulating support.

When an electrical current is passed through the conductive elements, the electroluminescent material emits light. EL displays, being an emissive technology, rather than shuttering a light source as per LCD displays, are most useful in applications where high visibility in all light conditions is important. The response speed for EL can be as fast as 5 1000 times that for LCD, thus making this mode particularly well suited for use with moving images.

Relatively recent developments in EL construction include devices comprising an organic luminescent medium consisting of two very thin layers. (< 1.0 $\mu$ m in combined thickness) 10 separating the anode and cathode. Representative of OLED devices are those disclosed in, for instance US 4720432.

The OLED devices have utilised a variety of cathode materials. Early investigations employed alkali metals. Other cathode materials taught by the art include combinations of 15 metals, such as brass and conductive metal oxides (e.g., indium tin oxide). A variety of single metal cathodes, such as indium, silver, tin, lead, magnesium, manganese, and aluminum, have also been used.

The present invention concerns the insulating and supporting substrate of an EL device, 20 specifically an OLED device such as a PLED device.

The substrates can be transparent, translucent or opaque, but are typically transparent. The substrates are usually required to meet stringent specifications for optical clarity, flatness and minimal birefringence. Typically, a total light transmission (TLT) of 85% over 400-25 800 nm coupled with a haze of less than 0.7% is desirable for displays applications. Surface smoothness and flatness are necessary to ensure the integrity of subsequently applied coatings such as the electrode conductive coating. The substrates should also have good barrier properties, i.e. high resistance to gas and solvent permeation. A substrate for use in electronic display applications suitably exhibits water vapour transmission rates of 30 less than 10<sup>-6</sup>g/m<sup>2</sup>/day and oxygen transmission rates of less than 10<sup>-5</sup>mL/m<sup>2</sup>/day. Mechanical properties such as flexibility, impact resistance, hardness and scratch resistance are also important considerations.

Optical quality glass or quartz has previously been used in electronic display applications as substrates. These materials are able to meet the optical and flatness requirements and have good thermal and chemical resistance and barrier properties. However, these materials do not have some of the desired mechanical properties, most notably low density, 5 flexibility and impact resistance.

In order to improve the mechanical properties, plastics materials have been proposed as replacements for glass or quartz sheet. One disadvantage of the use of polymeric materials is their lower chemical resistance and inferior barrier properties. Nevertheless, various 10 barrier coatings have been developed to minimise this problem. These coatings are typically applied in a sputtering process at elevated temperatures wherein the density and morphology of the coating can be controlled to give the required barrier properties. A barrier layer may be organic or inorganic, should exhibit good affinity for the layer deposited thereupon, and be capable of forming a smooth surface. Materials which are 15 suitable for use to form a barrier layer are disclosed, for instance, in US-6,198,217. In order to ensure the integrity of the barrier layer and to prevent "pin-pricks" therein, the surface of the polymeric substrate must exhibit good smoothness.

It is now possible to produce electronic display devices comprising barrier-coated 20 polymeric materials which have greater flexibility and improved impact resistance, and are of lighter weight than glass or quartz sheets of equal thickness. In addition, it is possible to provide polymeric substrates which exhibit good high-temperature dimensional stability, for instance during the high temperature techniques (such as sputtering) used to deposit the afore-mentioned barrier layer.

25 The surface smoothness of the polymeric substrate, however, is often inferior to conventional glass substrates. As noted previously, surface smoothness is critical in order to ensure the integrity of the subsequently applied barrier and conductive coatings, and to avoid pin-pricks

30 It is an object of this invention to provide a coated polymeric film substrate which overcomes at least one of the aforementioned problems. In particular, it is an object of this invention to provide a coated polymeric film substrate having improved surface

smoothness, particularly wherein said substrate is suitable for use in the manufacture of electroluminescent displays, particularly OLED devices.

As used herein, the term organic light emitting display (OLED) device refers to a display 5 device comprising a layer of light-emitting organic material (such as a polymer) disposed between two layers each of which comprises an electrode, wherein the resultant composite structure is disposed between two substrate (or support or cover) layers.

According to the present invention, there is provided the use of a composition comprising:

10 (a) about from 5 to 50 weight percent solids, the solids comprising about from 10 to 70 weight percent silica and about from 90 to 30 weight percent of a partially polymerized organic silanol of the general formula  $\text{RSi}(\text{OH})_3$ , wherein R is selected from methyl and up to about 40% of a radical selected from the group consisting of vinyl, phenyl, gamma-glycidoxypropyl, and gamma-methacryloxypropyl, and

15 (b) about from 95 to 50 weight percent solvent, the solvent comprising about from 10 to 90 weight percent water and about from 90 to 10 weight percent lower aliphatic alcohol, wherein the coating composition has a pH of about from 3.0 to 8.0, preferably from about 3.0 to about 6.5,

for the purpose of improving the surface smoothness of a polymeric substrate when applied 20 thereto, particularly wherein said use is in the manufacture of an electroluminescent display device, particularly an OLED device, which comprises a polymeric substrate.

Preferably, the pH of the coating composition is less than 6.2, preferably less than 6.0, 25 preferably less than 5.5, and preferably no more than 5.0. Preferably, the pH of the coating composition is at least 3.5, preferably at least 4.0. In one embodiment, the pH of the coating composition is in the range of from about 4.0 to about 5.0, preferably from about 4.1 to about 4.8, preferably from about 4.3 to about 4.5. The pH of the coating solution can be adjusted according to techniques well known to those skilled in the art, including the addition of an acidic or basic solution. For example, suitable acids for the adjustment of the 30 pH include hydrochloric and acetic acids, and suitable bases include sodium hydroxide.

The silica component of the coating compositions may be obtained, for example, by the hydrolysis of tetraethyl orthosilicate to form polysilicic acid. The hydrolysis can be carried

out using conventional procedures, for example, by the addition of an aliphatic alcohol and an acid. Alternatively, the silica used in the instant coating compositions can be colloidal silica. The colloidal silica should generally have a particle size of about from 5-25 millimicrons, and preferably about from 7-15 millimicrons. The alkalinity of the colloidal silica dispersions should generally be less than about 1%, and preferably less than 0.4% for optimum coatings and solution lifetime. Typical colloidal silicas which can be used in the instant invention include those commercially available from E. I. du Pont de Nemours and Company as "Ludox SM", "Ludox HS-30" and "Ludox LS" dispersions.

5       silica dispersions should generally be less than about 1%, and preferably less than 0.4% for optimum coatings and solution lifetime. Typical colloidal silicas which can be used in the instant invention include those commercially available from E. I. du Pont de Nemours and Company as "Ludox SM", "Ludox HS-30" and "Ludox LS" dispersions.

10      The organic silanol component of the instant composition has the general formula  $\text{RSi(OH)}_3$ . At least about 60% of the R groups, and preferably about from 80% to 100% of these groups, are methyl. Up to about 40% of the R groups can be higher alkyl or aryl selected from vinyl, phenyl, gamma-glycidoxypropyl, and gamma-methacryloxypropyl.

15      The combined solids of the coating composition, comprising the silica and the partially polymerized organic silanol, generally make up about from 5 to 50 weight percent of the total coating composition. Of these solids, the silica should comprise about from 10 to 70 weight percent, and preferably about from 20 to 60 weight percent, the complementary remainder comprising the organic siloxanol. Weight percents of the organic siloxanol 20 herein are calculated as  $\text{RSiO}_{1.5}$ .

The solvent component of the coating compositions generally comprises a mixture of water and one or more lower aliphatic alcohols. The water generally comprises about from 10 to 90 weight percent of the solvent, while the lower aliphatic alcohol complementarily 25 comprises about from 90 to 10 weight percent. The aliphatic alcohols generally are those having from 1 to 4 carbon atoms, such as methanol, ethanol, n-propanol, iso-propanol, n-butanol, sec-butanol and tertiary butanol.

In addition to the basic solvent components of water and alcohol, the solvent portion of the 30 compositions can further comprise up to about 10 weight percent of a compatible polar solvent such as acetone, ethylene glycol monoethylether, ethylene glycol monobutylether and diethylene glycol monoethylether.

Other suitable coating compositions are disclosed in US-5069942 and US-5425942, the disclosures of which are incorporated herein by reference. The compositions disclosed therein have a reduced alkali metal cation content and exhibit, *inter alia*, improved adhesion to a polymeric substrate.

5

Still further components which can be present in the coating compositions include curing catalysts. These are preferably present in a concentration of about from 0.01% to 0.1% based on the total weight of the composition, and especially about from 0.01 to 0.3 weight percent. Curing catalysts which may be used in the coating compositions can vary widely.

10 Representative catalysts include the alkali metal salts of carboxylic acids such as sodium acetate, potassium acetate, sodium formate, and potassium formate. Other representative curing catalysts which can be used include the quaternary ammonium carboxylates, such as benzyltrimethylammonium acetate.

15 The compositions can be prepared by wide variety of techniques, depending on the particular starting materials used. For example, organotrialkoxysilane can be hydrolyzed in the presence of prehydrolyzed polysilicic acid. Alternatively, organotrialkoxysilane can be prehydrolyzed and then added to a solution of polysilicic acid, often resulting in particularly rapid cure times. Still another alternative in the preparation of these  
20 compositions is the cohydrolysis of organotrialkoxysilane and tetraethyl orthosilicate together.

If a colloidal silica is used as the silica source in the coating compositions, the organic silanol can be combined with the silica either through the prehydrolysis of the  
25 organotrialkoxysilane or by hydrolyzing the organotrialkoxysilane in the presence of acidified colloidal silica dispersion. Still other methods of preparing and combining the components required for the compositions will be evident to those skilled in the handling of the individual components.

30 The coating compositions can be applied using conventional coating techniques, including continuous as well as dip coating procedures. The coatings are generally applied in thickness of about from 1 to 20 microns, and particularly about from 3 to 10 microns. The coating composition can be applied either "off-line" as a process step distinct from the fil-

manufacture, or "in-line" as a continuation of the film manufacturing process. In order to improve the surface smoothness of the coated film, it is desirable to avoid contamination from dust-particles and the like, and so the coating is preferably conducted off-line in a dust-free environment.

5

The coating compositions, after application to the substrate, can be cured at a temperature of about from 20 to 150°C. While ambient temperatures of 20°C require cure times of several days, elevated temperatures of 150°C will cure the coatings in several seconds. For convenience in commercial operations, a curing temperature of about 100°C is preferred, at 10 which cure times of about one minute are realised.

In a preferred embodiment, the substrate is a polyester film, such as poly(ethylene terephthalate) (PET) or poly(ethylene naphthalate) (PEN), preferably PEN, and particularly a heat-stabilised, heat-set oriented film comprising poly(ethylene naphthalate). In a 15 particularly preferred embodiment, the substrate is a heat-stabilised, heat-set, oriented film comprising poly(ethylene naphthalate) having a coefficient of linear thermal expansion (CLTE) within the temperature range from -40 °C to +100 °C of less than  $40 \times 10^{-6}/^\circ\text{C}$ , preferably less than  $30 \times 10^{-6}/^\circ\text{C}$ , more preferably less than  $25 \times 10^{-6}/^\circ\text{C}$ , more preferably less than 20  $\times 10^{-6}/^\circ\text{C}$ . Preferably the substrate has a shrinkage at 30 mins at 230°C, measured as 20 defined herein, of less than 1%, preferably less than 0.5%, and more preferably less than 0.1%. The preferred substrate and its preparation are described in more detail below.

The substrate is self-supporting by which is meant capable of independent existence in the absence of a supporting base. The thickness of the substrate is preferably between about 12 25 and 300  $\mu\text{m}$ , more preferably between about 25 and 250  $\mu\text{m}$ , more preferably between about 50 and 250  $\mu\text{m}$ .

PEN polyester can be synthesised by conventional methods. A typical process involves a direct esterification or ester exchange reaction, followed by polycondensation. Thus, PEN 30 polyester may be obtained by condensing 2,5-, 2,6- or 2,7-naphthalenedicarboxylic acid, preferably 2,6-naphthalenedicarboxylic acid, or a lower alkyl (up to 6 carbon atoms) diester thereof, with ethylene glycol. Typically, polycondensation includes a solid phase polymerisation stage. The solid phase polymerisation may be carried out on a fluidised

bed, e.g. fluidised with nitrogen, or on a vacuum fluidised bed, using a rotary vacuum drier. Suitable solid phase polymerisation techniques are disclosed in, for example, EP-A 0419400 the disclosure of which is incorporated herein by reference.

- 5 In a preferred embodiment, the PEN is prepared using germanium catalysts which provide a polymeric material having a reduced level of contaminants such as catalyst residue undesirable inorganic deposits and other byproducts of the polymer manufacture. As result of the "cleaner" polymeric composition, a film manufactured therefrom will exhibit improved optical clarity and surface smoothness.

10

- 15 The PEN used to prepare the substrate suitably has a PET-equivalent intrinsic viscosity (IV; measured as described herein) of 0.5-1.5, preferably 0.7-1.5, and in particular 0.7-1.0. An IV of less than 0.5 results in a polymeric film lacking desired properties such as mechanical properties whereas an IV of greater than 1.5 is difficult to achieve and would likely lead to processing difficulties of the raw material.

- 20 Formation of the substrate may be effected by conventional techniques well-known in the art. Conveniently, formation of the substrate is effected by extrusion, in accordance with the procedure described below. In general terms the process comprises the steps of extruding a layer of molten polymer, quenching the extrudate and orienting the quenched extrudate in at least one direction.

- 25 The substrate may be uniaxially-oriented, but is preferably biaxially-oriented. Orientation may be effected by any process known in the art for producing an oriented film, for example a tubular or flat film process. Biaxial orientation is effected by drawing in two mutually perpendicular directions in the plane of the film to achieve a satisfactory combination of mechanical and physical properties.

- 30 In a tubular process, simultaneous biaxial orientation may be effected by extruding a thermoplastics polyester tube which is subsequently quenched, reheated and then expanded by internal gas pressure to induce transverse orientation, and withdrawn at a rate which will induce longitudinal orientation.

In the preferred flat film process, the substrate-forming polyester is extruded through a slot die and rapidly quenched upon a chilled casting drum to ensure that the polyester is quenched to the amorphous state. Orientation is then effected by stretching the quenched  
5 extrudate in at least one direction at a temperature above the glass transition temperature of the polyester. Sequential orientation may be effected by stretching a flat, quenched extrudate firstly in one direction, usually the longitudinal direction, i.e. the forward direction through the film stretching machine, and then in the transverse direction. Forward stretching of the extrudate is conveniently effected over a set of rotating rolls or  
10 between two pairs of nip rolls, transverse stretching then being effected in a stenter apparatus. Alternatively, orientation may be generated in the extruded film by way of simultaneous stretching. Here, the film is stretched in the longitudinal and transverse directions in what is essentially the same stage of the process, in the stenter oven. For both routes of sequential and simultaneous stretching, the extent of stretching is determined  
15 partly by the nature of the polyester. However the film is usually stretched so that the dimension of the oriented film is from 2 to 5, more preferably 2.5 to 4.5 times its original dimension in each direction of stretching. Typically, stretching is effected at temperatures in the range of 70 to 140°C. Greater draw ratios (for example, up to about 8 times) may be used if orientation in only one direction is required. It is not necessary to stretch equally in  
20 the machine and transverse directions although this is preferred if balanced properties are desired.

The stretched film is dimensionally stabilised by heat-setting under dimensional restraint at a temperature above the glass transition temperature of the polyester but below the melting  
25 temperature thereof, to induce crystallisation of the polyester, as described in GB-A-838708. The tension of dimensional restraint is generally in the range of about 19 to about 75 kg/m, preferably about 45 to about 50 kg/m of film width which, for a film having a width of about 2.6 m is a tension in the range of about 50 to about 190 kg, preferably in the range of 120-130 kg. The actual heat-set temperature and time will vary depending on the  
30 composition of the film but should be selected so as not to substantially degrade the tear resistant properties of the film. Within these constraints, a heat-set temperature of about 135° to 250°C is generally desirable, more preferably 235-240°C. The duration of heating

will depend on the temperature used but is typically in the range of 5 to 40 secs, preferably 8 to 30 secs.

The film is then further heat-stabilised by heating it under low tension (i.e. with the minimum possible dimensional restraint) at a temperature above the glass transition temperature of the polyester but below the melting point thereof, in order to allow the majority of the inherent shrinkage in the film to occur (relax out) and thereby produce a film with very low residual shrinkage and consequently high dimensional stability. The tension experienced by the film during this heat-stabilisation step is typically less than 5 kg/m, preferably less than 3.5 kg/m, more preferably in the range of from 1 to about 2.5 kg/m, and typically in the range of 1.5 to 2 kg/m of film width. The temperature to be used for the heat stabilisation step can vary depending on the desired combination of properties from the final film, with a higher temperature giving better, i.e. lower residual shrinkage properties. A temperature of 135°C to 250°C is generally desirable, preferably 190 to 250°C, more preferably 200 to 230°C. The duration of heating will depend on the temperature used but is typically in the range of 10 to 40 sec, with a duration of 20 to 30 secs being preferred. This heat stabilisation process can be carried out by a variety of methods, including flat and vertical configurations and either "off-line" as a separate process step or "in-line" as a continuation of the film manufacturing process. In one embodiment, heat stabilisation is conducted "off-line".

The substrate may comprise one or more discrete layers. The composition of the respective layers may be the same or different. For instance, the substrate may comprise one, two, three, four or five or more layers and typical multi-layer structures may be of the AEB 25 ABA, ABC, ABAB, ABABA or ABCBA type. Preferably, the substrate comprises only one layer. Where the substrate comprises more than one layer, preparation of the substrates is conveniently effected by coextrusion, either by simultaneous coextrusion of the respective film-forming layers through independent orifices of a multi-orifice die, and thereafter uniting the still molten layers, or, preferably, by single-channel coextrusion in which molten streams of the respective polymers are first united within a channel leading to a die manifold, and thereafter extruded together from the die orifice under conditions of streamline flow without intermixing thereby to produce a multi-layer polymeric film which may be oriented and heat-set as hereinbefore described. Formation of a multi-layer

substrate may also be effected by conventional lamination techniques, for example by laminating together a preformed first layer and a preformed second layer, or by casting, for example, the first layer onto a preformed second layer.

5 The substrate may conveniently contain any of the additives conventionally employed in the manufacture of polymeric films. Thus, agents such as cross-linking agents, dyes, pigments, voiding agents, lubricants, anti-oxidants, radical scavengers, UV absorbers, thermal stabilisers, flame retardants, anti-blocking agents, surface active agents, slip aids, optical brighteners, gloss improvers, prodegradents, viscosity modifiers and dispersion  
10 stabilisers may be incorporated as appropriate. The components of the substrate may be mixed together in a conventional manner. For example, by mixing with the monomeric reactants from which the layer polymer is derived, or the components may be mixed with the polymer by tumble or dry blending or by compounding in an extruder, followed by cooling and, usually, comminution into granules or chips. Masterbatching technology may  
15 also be employed.

In a preferred embodiment, the film of the present invention is optically clear, preferably having a % of scattered visible light (haze) of <3.5%, preferably <2%, more preferably <1.5 %, more preferably ≤1%, and particularly less than 0.7%, measured according to the  
20 standard ASTM D 1003. Preferably the total light transmission (TLT) in the range of 400-800 nm is at least 75%, preferably at least 80%, and more preferably at least 85%, measured according to the standard ASTM D 1003. In this embodiment, filler is typically present in only small amounts, generally not exceeding 0.5% and preferably less than 0.2% by weight of a given layer.

25 In an alternative embodiment, the substrate is opaque and highly filled, preferably exhibiting a Transmission Optical Density (TOD) (Sakura Densitometer; type PDA 65; transmission mode) in the range from 0.1 to 2.0, more preferably 0.2 to 1.5, more preferably from 0.25 to 1.25, more preferably from 0.35 to 0.75 and particularly 0.45 to  
30 0.65. The film is conveniently rendered opaque by incorporation into the polymer blend of an effective amount of an opacifying agent. Suitable opacifying agents include an incompatible resin filler, a particulate inorganic filler or a mixture of two or more such fillers. Preferred particulate inorganic fillers include titanium dioxide and silica. Suitable

incompatible resins include polyamides and olefin polymers, particularly a homo- or co-polymer of a mono-alpha-olefin containing up to 6 carbon atoms in its molecule. The amount of filler present in a given layer is preferably in the range from 1% to 30%, more preferably 3% to 20%, particularly 4% to 15%, and especially 5% to 10% by weight, based  
5 on the weight of the layer polymer.

Prior to coating the substrate, it may be desirable to pretreat the substrate to promote adhesion of the coating. Various adhesion promoting techniques known to those skilled in the art can be used, such as flame treating, corona discharge, and/or resin coating.  
10

In a preferred embodiment, the substrate is coated with a primer layer to improve adhesion of the substrate to the planarising coating composition. In one embodiment, the primer layer is an acrylate or methacrylate polymer resin. Suitable materials include:

- (i) a copolymer of (a) 35 to 40 mole % alkyl acrylate, (b) 35 to 40 % alkyl  
15 methacrylate, (c) 10 to 15 mole % of a comonomer containing a free carboxyl group such as itaconic acid, and (d) 15 to 20 mole % of an aromatic sulphonic acid and/or salt thereof such as p-styrene sulphonic acid, an example of which is a copolymer comprising ethyl acrylate/methyl methacrylate/itaconic acid/p-styrene sulphonic acid and/or a salt thereof in a ratio of 37.5/37.5/10/15 mole %, as disclosed in EP-A-0429179 the disclosure of which  
20 is incorporated herein by reference; and
- (ii) an acrylic and/or methacrylic polymeric resin, an example of which is a polymer comprising about 35 to 60 mole % ethyl acrylate, about 30 to 55 mole % methyl methacrylate and about 2 to 20 mole % methacrylamide, as disclosed in EP-A-0408197 the disclosure of which is incorporated herein by reference.

25

The primer or adherent layer may also comprise a cross-linking agent which functions to cross-link the coating composition to improve adhesion to the substrate and should also be capable of internal cross-linking. Suitable cross-linking agents include optionally alkoxylated condensation products of melamine with formaldehyde. The primer or  
30 adherent layer may also comprise a cross-linking catalyst, such as ammonium sulphate, to facilitate the cross-linking of the cross-linking agent. Other suitable cross-linking agents and catalysts are disclosed in EP-A-0429179, the disclosures of which are incorporated herein by reference.

A further suitable primer is disclosed in US-3,443,950, the disclosure of which is incorporated herein by reference.

5 The coating of the primer layer onto the substrate may be performed in-line or off-line, but is preferably performed "in-line", and preferably between the forward and sideways stretches of a biaxial stretching operation.

Once the planarising coating composition has been coated onto the substrate, the coated 10 substrate is then ready for further processing and coating in preparation for its end-use. In the manufacture of an OLED device, the film is then coated with a barrier layer, as noted above. Such coatings are known in the art and are typically applied in a sputtering process at elevated temperatures. Materials which are suitable for use to form a barrier layer are disclosed, for instance, in US-6,198,217. An organic barrier layer may be formed from, for 15 instance, photocurable monomers or oligomers, or thermoplastic resins. Photocurable monomers or oligomers should have low volatility and high melting points. Examples of such monomers include trimethylol acrylates such as trimethylolpropane triacrylate, ditrimethylolpropane tetraacrylate and the like; long-chain acrylates such as 1,6-hexanediol diacrylate, 1,6-hexanediol dimethacrylate and the like; and cyclohexyl acrylates such as 20 dicyclopentyloxyethyl acrylate, dicyclopentyloxy acrylate, cyclohexyl methacrylate and the like. Examples of such oligomers include acrylate oligomers, epoxy acrylate oligomers, urethane acrylate oligomers, ether acrylate oligomers, and the like. Photoinitiators, such as benzoin ethers, benzophenones, acetophenones, ketals and the like, 25 may be used to cure the resin. Examples of suitable thermoplastic resins include polyethylene, polymethyl methacrylate, polyethylene terephthalate and the like. These organic materials may be applied by any conventional technique known in the art, such as by vacuum deposition. An inorganic barrier layer should be made of a material which exhibits low moisture permeability and is stable against moisture. Examples include oxides such as SiO<sub>2</sub>, SiO, GeO, Al<sub>2</sub>O<sub>3</sub> and the like, nitrides such as TiN, Si<sub>3</sub>N<sub>4</sub> and the like, and 30 metals such as Al, Ag, Au, Pt, Ni and the like. The inorganic material may be applied using a vapour phase technique such as vacuum deposition, sputtering and the like under standard conditions. Once the barrier layer has been applied, the substrate is ready for application of the conductive coatings which form the electrodes of the OLED.

According to a further aspect of the invention there is provided a coated, heat-stabilised, heat-set, oriented substrate comprising poly(ethylene naphthalate) having a coefficient of linear thermal expansion (CLTE) within the temperature range from -40 °C to +100 °C of less than  $40 \times 10^{-6}/^{\circ}\text{C}$ , preferably less than  $30 \times 10^{-6}/^{\circ}\text{C}$ , more preferably less than  $25 \times 10^{-6}/^{\circ}\text{C}$ , more preferably less than  $20 \times 10^{-6}/^{\circ}\text{C}$ , wherein the substrate preferably has a shrinkage at 30 mins at 230°C of less than 1%, preferably less than 0.5%, and more preferably less than 0.1%, wherein the coating is sufficient to improve the surface smoothness of said substrate, preferably such that the Ra value, as measured herein, is less than 0.6 nm, preferably less than 0.5 nm, preferably less than 0.4 nm, preferably less than 0.3 nm, and ideally less than 0.25 nm, and preferably such that the Rq value, as measured herein, is less than 0.8 nm, preferably less than 0.75 nm, preferably less than 0.65 nm, preferably less than 0.6 nm, preferably less than 0.50 nm, preferably 0.45nm or lower, preferably less than 0.35 nm, and ideally less than 0.3 nm. In a preferred embodiment of this aspect of the invention, the coating comprises a polysiloxane derived from a coating composition comprising:

- (a) about from 5 to 50 weight percent solids, the solids comprising about from 10 to 70 weight percent silica and about from 90 to 30 weight percent of a partially polymerized organic silanol of the general formula  $\text{RSi}(\text{OH})_3$ , wherein R is selected from methyl and up to about 40% of a radical selected from the group consisting of vinyl, phenyl, gamma-glycidoxypipropyl, and gamma-methacryloxypropyl, and
- (b) about from 95 to 50 weight percent solvent, the solvent comprising about from 10 to 90 weight percent water and about from 90 to 10 weight percent lower aliphatic alcohol, wherein the coating composition has a pH of about from 3.0 to 8.0.

25

According to a further aspect of the invention there is provided a method of preparing a coated polymeric film which comprises the steps of:

- (i) forming a layer comprising poly(ethylene naphthalate);
- (ii) stretching the layer in at least one direction;
- (iii) heat-setting under dimensional restraint at a tension in the range of about 19 to about 75 kg/m, preferably about 45 to about 50 kg/m of film width, at a temperature above the glass transition temperature of the polyester but below the melting temperature thereof;

(iv) heat-stabilising under low tension, preferably at a tension of less than 5 kg/m, more preferably at a tension of less than 3.5 kg/m, more preferably at a tension in the range of 1.0 to 2.5 kg/m, and typically at a tension in the range of 1.5 to 2.0 kg/m of film width, and at a temperature above the glass transition temperature of the polyester but below the melting temperature thereof; and

(v) applying a planarising coating composition thereto preferably such that the Ra value, as measured herein, is less than 0.6 nm, preferably less than 0.5 nm, preferably less than 0.4 nm, preferably less than 0.3 nm, and ideally less than 0.25 nm, and preferably such that the Rq value, as measured herein, is less than 0.8 nm, preferably less than 0.75 nm, preferably less than 0.65 nm, preferably less than 0.6 nm, preferably less than 0.50 nm, preferably 0.45nm or lower, preferably less than 0.35 nm, and ideally less than 0.3 nm..

According to a further aspect of the invention there is provided a coated, heat-stabilised, heat-set, oriented film comprising poly(ethylene naphthalate) obtainable by a method comprising the steps of:

- (i) forming a layer comprising poly(ethylene naphthalate);
- (ii) stretching the layer in at least one direction;
- (iii) heat-setting under dimensional restraint at a tension in the range of about 19 to about 75 kg/m, preferably about 45 to about 50 kg/m of film width, at a temperature above the glass transition temperature of the polyester but below the melting temperature thereof;
- (iv) heat-stabilising under low tension, preferably at a tension of less than 5 kg/m, more preferably at a tension of less than 3.5 kg/m, more preferably at a tension in the range of 1.0 to 2.5 kg/m, and typically at a tension in the range of 1.5 to 2.0 kg/m of film width, and at a temperature above the glass transition temperature of the polyester but below the melting temperature thereof; and
- (v) applying a planarising coating composition thereto preferably such that the Ra value, as measured herein, is less than 0.6 nm, preferably less than 0.5 nm, preferably less than 0.4 nm, preferably less than 0.3 nm, and ideally less than 0.25 nm, and preferably such that the Rq value, as measured herein, is less than 0.8 nm, preferably less than 0.75 nm, preferably less than 0.65 nm, preferably less than 0.6 nm, preferably less than 0.50 nm, preferably 0.45nm or lower, preferably less than 0.35 nm, and ideally less than 0.3 nm..

According to a further aspect of the present invention, there is provided a method for the manufacture of an electroluminescent display device, comprising the steps of:

- (i) forming a layer comprising poly(ethylene naphthalate);
- (ii) stretching the layer in at least one direction;
- 5 (iii) heat-setting under dimensional restraint at a tension in the range of about 19 to about 75 kg/m, preferably about 45 to about 50 kg/m of film width, at a temperature above the glass transition temperature of the polyester but below the melting temperature thereof;
- (iv) heat-stabilising under low tension, preferably at a tension of less than 5 kg/m, more preferably at a tension of less than 3.5 kg/m, more preferably at a tension in the range of
- 10 1.0 to 2.5 kg/m, and typically at a tension in the range of 1.5 to 2.0 kg/m of film width, and at a temperature above the glass transition temperature of the polyester but below the melting temperature thereof;
- (v) applying a planarising coating composition thereto preferably such that the Ra value, as measured herein, is less than 0.6 nm, preferably less than 0.5 nm, preferably less than 0.4 nm, preferably less than 0.3 nm, and ideally less than 0.25 nm, and preferably such that the Rq value, as measured herein, is less than 0.8 nm, preferably less than 0.75 nm, preferably less than 0.65 nm, preferably less than 0.6 nm, preferably less than 0.50 nm, preferably 0.45nm or lower, preferably less than 0.35 nm, and ideally less than 0.3 nm.; and
- 20 (vi) providing the coated, heat-stabilised, heat-set, oriented film as a substrate in the electroluminescent display device.

As used herein, the term "planarising coating composition" refers to a polymeric coating composition which increases the surface smoothness of a substrate when applied thereto, preferably such that the surface smoothness is improved such that the Ra value, as measured herein, is less than 0.6 nm, preferably less than 0.5 nm, preferably less than 0.4 nm, preferably less than 0.3 nm, and ideally less than 0.25 nm, and preferably such that the Rq value, as measured herein, is less than 0.8 nm, preferably less than 0.75 nm, preferably less than 0.65 nm, preferably less than 0.6 nm, preferably less than 0.50 nm, preferably 0.45nm or lower, preferably less than 0.35 nm, and ideally less than 0.3 nm. In a preferred embodiment of this aspect of the invention, the planarising coating composition comprises a polysiloxane derived from a composition comprising:

(a) about from 5 to 50 weight percent solids, the solids comprising about from 10 to 70 weight percent silica and about from 90 to 30 weight percent of a partially polymerized organic silanol of the general formula  $\text{RSi}(\text{OH})_3$ , wherein R is selected from methyl and up to about 40% of a radical selected from the group consisting of vinyl, phenyl, gamma-glycidoxypipropyl, and gamma-methacryloxypropyl, and

5 (b) about from 95 to 50 weight percent solvent, the solvent comprising about from 10 to 90 weight percent water and about from 90 to 10 weight percent lower aliphatic alcohol, wherein the coating composition has a pH of about from 3.0 to 8.0.

10 According to a further aspect of the invention, there is provided the use of a planarising coating composition in the manufacture of an electroluminescent display device, particularly an OLED device, which comprises a polymeric substrate, for the purpose of improving the surface smoothness of said polymeric substrate.

15 The following test methods may be used to determine certain properties of the polymeric film:

(i) the clarity of the film may be evaluated by measuring total luminance transmission (TLT) and haze (% of scattered transmitted visible light) through the total thickness of the film using a Gardner XL 211 hazemeter in accordance with ASTM D-1003-

20 61.

(ii) Transmission Optical Density (TOD) of the film may be measured using a Macbeth Densitometer TR 927 (Dent & Woods Ltd, Basingstoke, UK) in transmission mode.

(iii) Dimensional stability may be assessed in terms of either (a) the coefficient of linear thermal expansion (CLTE) or (b) a temperature cycling method wherein the residual change in length along a given axis is measured after heating the film to a given temperature and subsequently cooling the film to room temperature.

25 Both methods of measurements were conducted using a Thermomechanical Analyser PE-TMA-7 (Perkin Elmer) calibrated and checked in accordance with known procedures for temperature, displacement, force, eigendeformation, baseline and furnace temperature alignment. The films were examined using extension analysis clamps. The baseline required for the extension clamps was obtained

30 using a very low coefficient of expansion specimen (quartz) and the CLTE

precision and accuracy (dependent on post-scan baseline subtraction) was assessed using a standard material, e.g. pure aluminium foil, for which the CLTE value is well known. The specimens, selected from known axes of orientation within the original film samples, were mounted in the system using a clamp separation of approx.12mm and subjected to an applied force of 75mN over a 5mm width. The applied force was adjusted for changes in film thickness, i.e. to ensure consistent tension, and the film was not curved along the axis of analysis. Specimen lengths were normalised to the length measured at a temperature of 23°C.

In the CLTE test method (a), specimens were cooled to 8°C, stabilised, then heated at 5°C/min from 8°C to +240°C. The CLTE values ( $\alpha$ ) were derived from the formula:

$$\alpha = \Delta L / (L \times (T_2 - T_1))$$

where  $\Delta L$  is the measured change in length of the specimen over the temperature range ( $T_2 - T_1$ ), and  $L$  is the original specimen length at 23°C. CLTE values are considered reliable up to the temperature of the  $T_g$  (120°C).

The data can be plotted as a function of the % change in specimen length with temperature, normalised to 23°C.

In the temperature cycling test method (b), a procedure similar to that of method (a) was used wherein the temperature was cycled between 8°C and several elevated temperatures. Thus, film samples were heated from 8°C to 140°C, 160°C, 180°C or 200°C and then cooled to 8°C. The length along each of the transverse and machine directions was measured at 25°C before and after this heat treatment and the change in length  $\Delta L_r$  calculated as percentage of the original length. The results are shown in Table 2.

#### 25 (iv) Intrinsic Viscosity (IV)

The IV was measured by melt viscometry, using the following procedure. The rate of flow pre-dried extrudate through a calibrated die at known temperature and pressure is measured by a transducer which is linked to a computer. The computer programme calculates melt viscosity values ( $\log_{10}$  viscosity) and equivalent IVs from a regression equation determined experimentally. A plot of the IV against time in minutes is made by the computer and the degradation rate is calculated. An extrapolation of the graph to zero time gives the initial IV and equivalent melt

viscosity. The die orifice diameter is 0.020 inches, with a melt temperature of 284°C for IV up to 0.80, and 295°C for IV>0.80.

(v) Shrinkage

Shrinkage at a given temperature is measured by placing the sample in a heated oven for a given period of time. The % shrinkage is calculated as the % change of dimension of the film in a given direction before and after heating.

(vi) Surface Roughness

Surface smoothness was measured using conventional non-contacting, white-light, phase-shifting interferometry techniques, which are well-known in the art. The instrument used was a Wyko NT3300 surface profiler. Useful characterising data obtainable using the technique include:

*Roughness Average (Ra) : the arithmetic average peak height calculated over the measured surface area.*

*Root Mean Square Roughness (Rq) : the root mean square average peak height calculated over the measured surface area.*

*Maximum Profile Peak Height (Rp) : the height of the highest peak in the measured surface area.*

*Average Maximum Profile Peak Height (Rpm) : the average value of the ten highest peaks in the measured surface area.*

The roughness parameters and peak heights are measured relative to the average level of the sample surface area, or "mean line", in accordance with conventional techniques. (A polymeric film surface may not be perfectly flat, and often has gentle undulations across its surface. The mean line is a straight line that runs centrally through undulations and surface height departures, dividing the profile such that there are equal areas above and below the mean line.)

The surface profile analysis is conducted by scanning discrete regions of the film surface within the "field of view" of the surface profiler machine, which is the area scanned in a single measurement. A film sample may be analysed using a discrete field of view, or by scanning successive fields of view to form an array. The analyses conducted herein utilised the full resolution of the Wyko NT3300 surface profiler, in which each field of view comprises 736 x 480 pixels.

For the measurement of Ra and Rq, the resolution was enhanced using an objective lens having a 50-times magnification. The resultant field of view has dimensions of 90 µm x 120 µm, with a pixel size of 0.163 µm.

5 For the measurement of Rp and Rpm, the resolution is conveniently enhanced using an objective lens having a 10-times magnification in combination with a "0.5-times field of view of multiplier" to give a total magnification of 5-times. The resultant field of view has dimensions of 0.9 mm x 1.2 mm, with a pixel size of 1.63 µm.

For each measurement, the results of five successive scans are combined to give an average value. The measurements were conducted using a modulation threshold

10 (signal : noise ratio) of 10%, i.e. data points below the threshold are discarded.

It is preferred that the films have an Ra value, as measured herein, of less than 0.7 nm, preferably less than 0.6 nm, preferably less than 0.5 nm, preferably less than 0.4 nm, preferably less than 0.3 nm, and ideally less than 0.25 nm. It is preferred that the films of the present invention have an Rq value, as measured herein, of less

15 than 0.9 nm, preferably less than 0.8 nm, preferably less than 0.75 nm, preferably less than 0.65 nm, preferably less than 0.6 nm, preferably less than 0.50 nm, preferably 0.45nm or lower, preferably less than 0.35 nm, and ideally less than 0.3 nm.

20 The invention is further illustrated by the following examples. It will be appreciated that the examples are for illustrative purposes only and are not intended to limit the invention as described above. Modification of detail may be made without departing from the scope of the invention.

25 **EXAMPLES**

**Preparation of Polyester A**

Dimethyl naphthalate was reacted with ethylene glycol in the presence of 400 ppm manganese acetate tetrahydrate catalyst to give bis-(2-hydroxyethyl) naphthalate and low  
30 oligomers thereof, in a standard ester interchange reaction. At the end of the ester interchange reaction 0.025% of phosphoric acid stabiliser was added, followed by 0.04% of antimony trioxide polycondensation catalyst. A standard batch polycondensation

reaction was performed until the intrinsic viscosity (IV) of the polyethylene naphthalate was approximately 0.50 - 0.55 (true PEN IV; PET equivalent IV 0.75 - 0.80)

Preparation of Polyester B

Dimethyl naphthalate was reacted with ethylene glycol (2.1:1 glycol:ester mole ratio) in 5 the presence of 400 ppm manganese acetate catalyst to give bis-(2-hydroxyethyl) naphthalate and low oligomers thereof, in a standard ester interchange reaction. At the end of the ester interchange reaction 0.025% of phosphoric acid stabiliser was added, followed by 0.020% of germanium dioxide polycondensation catalyst (133ppm Ge metal). A standard batch polycondensation reaction was performed until the intrinsic viscosity 10 (IV) of the polyethylene naphthalate was approximately 0.50 - 0.55 (true PEN IV; PET equivalent IV 0.75 - 0.80).

EXAMPLES 1 and 2

A polymer composition comprising PEN was extruded and cast onto a hot rotating 15 polished drum. The film was then fed to a forward draw unit where it was stretched over a series of temperature-controlled rollers in the direction of extrusion to approximately 3.34 times its original dimensions. The draw temperature was approximately 133°C. The film was then passed into a stenter oven at a temperature of 138°C where the film was stretched in the sideways direction to approximately 4.0 times its original dimensions. The biaxially 20 stretched film was then heat-set at temperatures up to about 238°C by conventional means before being cooled and wound onto reels. The total film thickness was 125 µm.

The heat-set biaxially stretched film was then unwound and passed through a series of four flotation ovens and allowed to relax by applying the minimum line tension compatible with 25 controlling the transport of the web. The heat-stabilised film was then wound up. Each of the four ovens had three controlled temperature zones in the transverse direction (left, centre and right):

	Left	Centre	Right
30 Oven 1	200	213	200
Oven 2	200	213	200
Oven 3	200	213	200
Oven 4	195	213	195

The line speed of the film during the heat-stabilisation step was 15m/min. The tensions used for the film (1360mm original roll width) were 24-25N.

5 The film of Example 1 was prepared using polyester A. The film of Example 2 was prepared using polyester B. Surface roughness data are in Table 1.

EXAMPLE 3

The film of Example 2 was coated, between the forwards and sideways stretches during  
10 film manufacture, with a first coating composition comprising  
(i) an 18% solids aqueous dispersion of a copolymer of ethyl acrylate (EA; 48 mole %),  
methyl methacrylate (MMA; 48 mole %) and methacrylamide (MA; 4 mole %) (derived  
from AC201 ®; Rohm and Haas); 18 litres  
(ii) SYNPERONIC NP10 ® (Uniqema; a nonyl phenol ethoxylated surfactant; 100ml  
15 (iii) 20% ammonium nitrate (20% aqueous solution); 300ml  
(iv) Distilled water; 81 litres.

The dry thickness of the primer coating was 30nm.

20 The film was then coated off-line with a second coating composition obtained as follows:  
(i) 517 cm<sup>3</sup> of methyltrimethoxysilane (obtained from OSi Specialities) was added to 1034  
cm<sup>3</sup> demineralised water at room temperature and stirred for 24 hours.  
(ii) 54 cm<sup>3</sup> of 3-glycidoxypropyl trimethoxysilane (obtained from Aldrich Chemical  
Company) was added to 108 cm<sup>3</sup> of demineralised water at room temperature and stirred  
25 for 24 hours.  
(iii) 53 cm<sup>3</sup> of 10 % aqueous acetic acid (Aldrich Chemical Company) was added to  
700cm<sup>3</sup> of Ludox LS colloidal silica (12 nm). To this was added 162 cm<sup>3</sup> of the hydrolysed  
3-glycidoxypropyl trimethoxysilane/water mixture and 1551cm<sup>3</sup> of the hydrolysed  
methyltrimethoxysilane/water mixture. This mixture was stirred for 12 hours before  
30 coating. The final pH of the composition was 4.4. The thickness of the coating was 2 µm.

The surface roughness of the coated film is shown in Table 1.

**Table 1. Surface Roughness**

	Ex. 1	Ex. 2	Ex. 3
Average Roughness (Ra)	0.64	0.63	0.58
Root Mean Square Roughness (Rq)	0.90	0.82	0.74

The results in Table 1 show that a superior smoothness is obtained for Example 3.

**CLAIMS**

1. The use of a composition comprising:
  - (a) about from 5 to 50 weight percent solids, the solids comprising about from 10 to 70 weight percent silica and about from 90 to 30 weight percent of a partially polymerized organic silanol of the general formula  $RSi(OH)_3$ , wherein R is selected from methyl and up to about 40% of a radical selected from the group consisting of vinyl, phenyl, gamma-glycidoxypropyl, and gamma-methacryloxypropyl, and
  - (b) about from 95 to 50 weight percent solvent, the solvent comprising about from 10 to 90 weight percent water and about from 90 to 10 weight percent lower aliphatic alcohol,  
wherein the coating composition has a pH of about from 3.0 to 8.0,  
for the purpose of improving the surface smoothness of a polymeric substrate when applied thereto.
- 15 2. The use of claim 1 wherein said substrate is a polyester film.
3. The use of claim 1 wherein said substrate is a poly(ethylene naphthalate) film.
- 20 4. The use of claim 2 or 3 wherein said substrate is a heat-stabilised, heat-set, oriented film.
5. The use of claim 2 or 3 wherein said substrate is a heat-stabilised, heat-set, oriented film comprising poly(ethylene naphthalate) having a coefficient of linear thermal expansion (CLTE) within the temperature range from -40 °C to +100 °C of less than  $40 \times 10^{-6}/^{\circ}C$ .
- 25 6. A use according to any preceding claim wherein said heat-stabilised film has shrinkage at 30 mins at 230°C of less than 1%.
- 30 7. A use according to any preceding claim wherein the poly(ethylene naphthalate) is derived from 2,6-naphthalenedicarboxylic acid.

8. A use according to any preceding claim wherein the poly(ethylene naphthalate) has an intrinsic viscosity of 0.5 – 1.5.
9. A use according to any preceding claim wherein said heat-stabilised film has a % of  
5 scattered visible light (haze) of <1.5%.
10. A use according to any preceding claim wherein said heat-stabilised film is biaxially oriented.

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